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Classical dynamics of laser-driven H_3^+

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[**Introduction**] To theoretically model the outcome of an experiment in which a molecule is subjected to a short and intense laser pulse [1,2], one is faced with several difficulties. In principle, the time-dependent Schrödinger equation (TDSE) should be solved. Since both electronic and nuclear dynamics proceed in parallel, and are strongly coupled, the theoretical simulation has to include both electronic and nuclear degrees of freedom. Typically, ionization of one or several electrons is followed by fragmentation into one of several possible pathways. The calculation must go beyond the approximation of frozen nuclei, so that we can calculate experimentally measured quantities such as the kinetic energy release. However, despite the tremendous progress both in computer power and numerical algorithms, the TDSE with an external, intense laser field including nuclear and electronic motion has so far only been solved for one-electron systems. If the nuclei are kept fixed, two electrons may be treated. To be able to simulate larger molecules, approximate schemes [3] or different models have to be employed.

In this contribution, we show how to model laser-molecule interaction with a semi-classical model. The idea is to treat both nuclei and electrons as classical point particles, and the dynamical evolution of the system by the classical equations of motion. In this way, numerical integration is easily performed, and experimentally observable quantities are straightforwardly calculated by averaging over many trajectories originating from the initial distribution of positions and momenta. Motivated by a recent experiment [2], we take the H_3^+ molecule as a test case to assess the ability of the model to describe strong-field-molecule interaction.

[**Theoretical model**] A classical molecule, with Coulombic inter-particle interaction, is not stable. Such a molecule will autodissociate, autoionize, or even collapse. In order to stabilize the field-free molecular structure, we extend the model in [4] to the H_3^+ molecule. The crucial ingredient is to add, in addition to the usual Coulomb terms, a repulsive, momentum-dependent potential $\phi(\boldsymbol{p},\boldsymbol{r})$ to the Hamiltonian H. The role of this potential is to implement the Heisenberg principle $|\boldsymbol{p}||\boldsymbol{r}| > 1$ (in atomic units) in an approximate way, by keeping the electrons from visiting parts of the phase space that would not be accessible in quantum mechanics. The Hamiltonian defining the H_3^+ system with three protons (momenta $\boldsymbol{P}_j$, positions $\boldsymbol{R}_j$, $1 \geq j \geq 3$, and mass M) and two electrons (momenta $\boldsymbol{p}_k$, positions $\boldsymbol{r}_k$, $1 \geq k \geq 2$) then reads

$$H = \sum_{j=1}^{3} \frac{\boldsymbol{P}_j^2}{2M} + \sum_{k=1}^{2} \frac{\boldsymbol{p}_k^2}{2} + \frac{1}{|\boldsymbol{r}_1 - \boldsymbol{r}_2|} + \sum_{j<k} \frac{1}{|\boldsymbol{R}_j - \boldsymbol{R}_k|} + \sum_{j=1}^{3}\sum_{k=1}^{2}\left[-\frac{1}{|\boldsymbol{s}_{jk}|} + \frac{\phi(\boldsymbol{q}_{jk},\boldsymbol{s}_{jk})}{\boldsymbol{s}_{jk}^2}\right] + \Phi(\boldsymbol{P}_{1,2,3},\boldsymbol{R}_{1,2,3},\boldsymbol{p}_{1,2},\boldsymbol{r}_{1,2}). \quad (1)$$

In Eq. (1), we used the notation $\boldsymbol{s}_{jk} = \boldsymbol{R}_j - \boldsymbol{r}_k$, and the relative momentum $\boldsymbol{q}_{jk} = (\boldsymbol{P}_j - M\boldsymbol{p}_k)/(M+1)$. The form actually used for the auxiliary potential ϕ reads $\phi(\boldsymbol{p},\boldsymbol{r}) = \exp\{4[1-(|\boldsymbol{p}||\boldsymbol{r}|)^4\}/16$. The last term Φ in Eq. (1) contains certain 3-, 4-, and 5-body potentials included to fine-tune the model, so that the minimum energy configurations of H_2^+, H_2, and H_3^+ match the accurate, quantum mechanical ones as closely as possible.

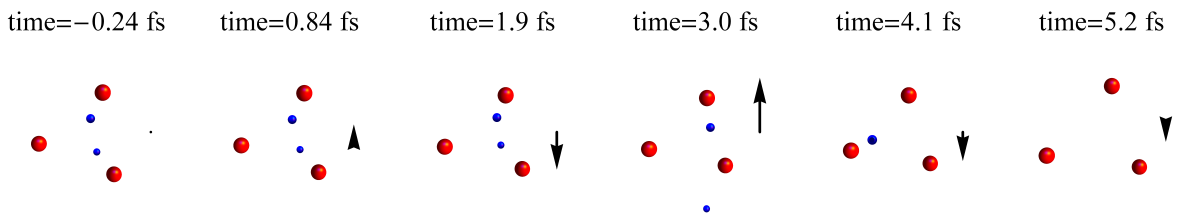

Figure 1: Snapshots of double ionization of the H_3^+ molecule by a 3-cycle laser pulse (pulse length 4 fs). Protons are shown as red, and electrons as blue spheres. The laser field vector is depicted with a black arrow, and is turned on at time = 0. After the ejection of both electrons by the strong laser pulse, the remaining protons will separate from each other by Coulomb explosion.

[**Results and discussion**] The classical equations of motion derived from the Hamiltonian (1), including the force induced by the laser pulse, were integrated numerically for a large number of slightly different initial values of the particle positions and momenta. The laser carrier wavelength of the linearly polarized 3-cycle pulse was chosen to be 790 nm. In the simulation, both ionization as well as dissociation pathways are seen, and all the final reaction products observed in the experiment are actually produced. In order to determine the intensity dependence of ionization and dissociation probabilities, a range of laser intensities were investigated. A typical trajectory resulting from the interaction with a highly intense laser pulse of intensity 4×10^{15} W/cm^2 is shown in Fig. 1. Here, both electrons are sequentially removed from the molecule at the laser field maxima, leaving only the three bare nuclei, which will subsequently fly apart due to the repulsive Coulomb force. We have also calculated the spectra of the kinetic energy release and different angular distributions, which show qualitative agreement with the experimental data [2].

We aim to extend the model to larger, hydrocarbon molecules. This model may thus provide a way, alternative to wave function-based methods, of getting an insight into laser-molecule interaction for larger systems than diatomic molecules.

[**References**]

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